

# 8 DAC #

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

**Masaui TAKAHASHI et al**

Atty. Ref.: **1858-23**

Serial No. **09/702,828**

Group: **1714**

Filed: **November 1, 2000**

Examiner: **(Unknown)**

For: **AUTOMOBILE INTERIOR OR EXTERIOR TRIM MATERIAL**

\* \* \* \* \*

10/08/2002 HBERHE 00000021 09702828

01 FC:128

1960.00 DP

October 4, 2002

Honorable Commissioner of Patents  
and Trademarks  
Washington, DC 20231

RECEIVED  
OCT 08 2002  
OFFICE OF PETITIONS

**SECOND RENEWED PETITION UNDER 37 CFR §§ 1.42 AND 1.47**

Sir:

This paper is being filed responsive to the "Decision Refusing Status Under 37 CFR 1.47(a)" dated March 18, 2002, which set a nominal response due date of May 18, 2001. Pursuant to 37 CFR 1.136(a), applicants hereby petition for a time extension from such nominal due date for five (5) months up to, and including, October 18, 2002. The appropriate extension fee in the amount of \$1,960.00 is attached.<sup>1</sup>

Applicants again renew their Petition pursuant to 37 CFR §1.42 and 1.47(a) to accept the above-identified application for examination in the absence of the signature of one of the joint inventors, namely Masayuki Takahashi, who is deceased and whose heiress-in-law cannot be located. In this regard, the Petition filed on April 11, 2001 as

<sup>1</sup> The Commissioner is hereby authorized to charge any fee deficiency deemed necessary for the consideration of this paper to the undersigned's Deposit Account No. 14-1140.

well as the Renewed Petition filed on December 18, 2001 are expressly incorporated hereinto by reference.

The Petitions Examiner has criticized the previous factual submissions on essentially three grounds, namely that (i) facts have not been proffered which establish that the deceased inventor's wife and legal representative as his heiress-in-law, Ms. Yuko Takahashi, has been presented with a complete copy of the subject application papers, (ii) a declaration in compliance with Rule 63 which includes Ms. Takahashi's last known address information has not been submitted, and (iii) a clear statement of Ms. Takahashi's last known address has not been submitted.

As to (i) above, there is attached hereto as Tab A a "Second Supplemental Factual Declaration In Support of Petition Under Rule 42 and 47" executed by Mr. Hideo Nakano (hereinafter the Second Supplemental Nakano Declaration"). As stated therein, a registered letter was sent to Ms. Yuko Takahashi on August 1, 2002 along with a complete copy of the subject application papers (i.e., an Abstract together with twenty six (26) pages of specification and claims, including specification page 1A, and also including 14 numbered claims). A Rule 63 Declaration form accompanied such complete application. To date, however, there has been no reply from Ms. Takahashi. As such, it must be concluded that Ms. Takahashi has constructively refused to join in the subject application.

As to (ii) above, there is attached hereto as Tab B a Supplemental Declaration which includes Ms. Takahashi's currently known address and which has been executed by the other cooperating joint inventors – namely, Messrs. Yukino and Fukushima.

As to (iii) above, the Petitions Examiner's attention is directed to paragraph 3 in the attached Second Supplemental Nakano Declaration. In the event that that paragraph is not a sufficiently clear statement of Ms. Takahashi's last known address as of this date, such address is reproduced below and should therefor be considered as such a statement of last known address:

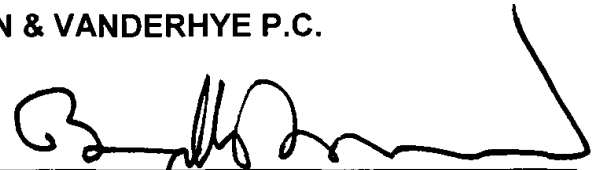
**Yuko Takahashi  
Raionzu-Manshion Wakaba  
Fuji Kou n 13-1-301  
Fujimi 2-chome  
Tsurugashima-shi Saitama 350-2201  
JAPAN**

It is believed that the totality of the factual evidence now of record makes it quite clear that Rule 47 status should be accorded to the present application. Such favorable action is therefore solicited.

Respectfully submitted,

**NIXON & VANDERHYE P.C.**

By: \_\_\_\_\_

A handwritten signature in black ink, appearing to read 'Bryan H. Davidson', written over a horizontal line.

Bryan H. Davidson  
Reg. No. 30,251

BHD:fmh  
1100 North Glebe Road, 8th Floor  
Arlington, VA 22201-4714  
Telephone: (703) 816-4000  
Facsimile: (703) 816-4100



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

Masayuki TAKAHASHI et al

Atty. Ref.: 1858-23

Serial No. 09/702,828

Group: 1714

Filed : November 1, 2000

Examiner: (Unknown)

For: AUTOMOBILE INTERIOR OR EXTERIOR TRIM  
MATERIAL

\* \* \* \* \*

Honorable Commissioner of Patents  
and Trademarks  
Washington, DC 20231

RECEIVED  
OCT 08 2002  
OFFICE OF PETITIONS

SECOND SUPPLEMENTAL FACTUAL DECLARATION IN  
SUPPORT OF PETITION UNDER RULES 42 AND 47

Sir:

The undersigned, Hideo NAKANO, hereby declares and states that:

1. I am the same individual who executed on April 4, 2001, the "Factual Declaration In Support of Petition Under Rules 42 and 47" (hereinafter "the original Factual Declaration") and executed on December 12, 2001, the "Supplemental Factual Declaration in Support of Petition Under Rules 42 and 47" (hereinafter "the supplemental Factual Declaration") in connection with the above-identified matter. All facts stated in the original and supplemental Factual Declarations are hereby incorporated expressly by reference as if the same were set forth herein.

2. On information and belief, I understand the United States Patent and Trademark Office has again criticized the facts submitted with the supplemental Factual Declaration as being insufficient to establish that a true and accurate copy of the entire application in this matter was sent and received by heiress-in-law, Ms. Yuko Takahashi, of the deceased (and thereby non-signing) inventor, Mr. Masayuki Takahashi.
3. In order to address such criticism, under my direction and control, on August 1, 2002, I sent a registered letter to Ms. Yuko Takahashi at her last known address, namely: Raionzu-Mansion Wakaba Fujimi Kouen 13-1-301, Fujimi 2-chome, Tsurugashima-shi, Saitama 350-2201 Japan. A copy of the registered letter and a partial translation into English-language thereof is attached hereto as Exhibit AA and a copy of the signed registered mail receipt and an English-language translation thereof indicating that Ms. Yuko Takahashi, or a person concerned, actually received the letter and its contents is attached hereto as Exhibit BB.
4. Enclosed with the letter noted in paragraph 3 above was a complete copy of the subject application comprising an Abstract together with twenty six (26) pages of specification and claims, including specification page 1A, and also including 14 numbered claims. An inventors' Declaration form in accordance with USPTO Rule 63 was attached to the complete application for Ms. Yuko Takahashi's execution, should she decide to join in the same.
5. As of this date, there has been no reply from Ms. Yuko Takahashi to the registered letter identified above in paragraph 3 above. I therefore must conclude that Ms. Yuko Takahashi has refused to join in the subject application in her representative capacity as heiress-in-law of her deceased inventor husband's estate.
6. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are

punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully Submitted,

ASAHI DENKA KOGYO K.K.

October 1, 2002

Date Signed

Hideo Nakano

By: Hideo NAKANO

Title: Manager of Patent & Information Section-Urawa  
Planning Department, R & D Division

1-1

2002年08月01日

埼玉県鶴ヶ島市富士見2丁目13番1号  
 ライオンズマンション若葉富士見公園301  
 高橋 祐子 様

旭電化工業株式会社  
 研究企画部 情報特許室  
 浦和情報特許 G (中野)  
 課長 中野 栄郎

拝啓、

高橋家の皆様方におかれましては益々ご健勝のこととお喜び申し上げます。

1-2

さて、先日ご依頼いたしましたアメリカ特許庁へ提出する書類への、サインと日付の記入の件、アメリカ特許庁より特許明細書を添付の上、再度連絡して意思を確認するようにとの指示がありました。

下記書類をお送り致します。

お忙しいところ恐れ入りますが、特許出願上必要な書類であり、諸般の事情をご賢察の上、ご理解頂きたく、宜しくお願い致します。

もし、ご不明な点がありましたら下記連絡先までお手紙、お電話を頂きますようお願い致します。

また、本状と行き違いのせつはご容赦下さい。

1-3

敬具

#### 同封書類

「自動車内外装材」に関わる書類として

日本出願公開公報 特開平11-310667号	1通
アメリカ出願英文明細書	1通
委任状	1通
譲渡証	1通

「造核剤」に関わる書類として

日本出願公開公報 特開2001-059040号	1通
アメリカ出願英文明細書	1通
委任状	1通
譲渡証	1通

連絡先：本件に関するご質問がありましたら、以下の連絡先へお願い致します。

旭電化工業株式会社

埼玉県さいたま市白幡5-2-13

研究企画部 情報特許室 浦和情報特許G

担当：(室長) 山田和正 (課長) 中野栄郎

電話：048-838-2222

FAX：048-838-2220

E-mail：hideo-n@adk.co.jp

## 書留・配達記録郵便物受領証 (お客様控)

(差出人の住所氏名)		さいたま市白幡5丁目2番13号 旭電化工業株式会社 樹脂添加剤開発研究所			様
受取人の氏名	引受番号	郵便料	申出損害賠償額	摘要	
高橋祐子 様	438-49- 17506-4	740	---	簡易	

ご注意 この受領証は、損害賠償の請求をするときその他の場合に必要ですから大切に保存してください。  
簡易書留の損害賠償額は、8千円を限度とする実損額です。

摘要欄：カン(簡易)、キロ(配達記録)、ソク(速達)、ハイ(配達証明)の記号 ナイ(内容証明)、トク(特別送達)、ダイ(代金引換)  
ジ(引受時刻証明)、シデ(配達日指定)

配達状況がわかります。フリーダイヤル 0120-232886  
インターネット <http://www.postal.mpt.go.jp/>  
ユ07150

浦和中央 郵便局

02.08.02\*12-18

Partial translation of the exhibits into English-language

Exhibit AA

1 - 1.

August 1, 2002

Ms. Yuko TAKAHASHI  
Raionzu-Manshion Wakaba Fujimi Kouen 301,  
13-1, Fujimi 2-chome, Tsurugashima,

Hideo NAKANO  
Manager of Patent & Information  
Section-Urawa Planning Department, R&D Division  
ASAHI DENKA KOGYO K.K,

1 - 2.

In connection with our request for your signature and date to the formal documents for filing a patent application to USPTO, we have been recently suggested from USPTO to confirm again your intention to execute the documents for the subject applications with accompanying English version specifications. I attached the formal documents for your signature and date listed below.

I am sorry to trouble you, but I hope you will understand the circumstances which are related to patent applications.

1 - 3.

Attached documents

A copy of Japanese Patent Application (lade open) No. HEI 11 -310667.

A copy of an English version of the corresponding Specification for U.S application.

An original Declaration and Power of attorney for signature.

An original Assignment for signature.

Exhibit BB

2 - 1.

Registered mail receipt (Duplicate for sender)

(Sender's address and name)

2-23, Shirahata 5-chome, Saitama-shi  
ASAHI DENKA KOGYO K.K,

Developmental Institute of Additives for resins

(Receiver's name)

Ms. Yuko TAKAHASHI

2 - 2.

URAWA central post office

August 2, 2002



RECEIVED

OCT 08 2002

1858-23  
FAS-170USNixon & Vanderhye P.C. (10/99)  
OFFICE OF PETITIONS  
(Domestic Non-Assigned/Foreign) Page 1RULE 63 (37 C.F.R. 1.63)  
INVENTORS DECLARATION FOR PATENT APPLICATION  
IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

As a below named inventor, I hereby declare that my residence, post office address and citizenship are as stated below next to my name, and I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

AUTOMOBILE INTERIOR OR EXTERIOR TRIM MATERIAL

the specification of which (check applicable box(es)):

☐ is attached hereto  
☒ was filed on November 1, 2000 as U.S. Application Serial No. 09/702,828 (Atty Dkt. No. 1858-23)  
☐ was filed as PCT International application No. \_\_\_\_\_ on \_\_\_\_\_  
and (if applicable to U.S. or PCT application) was amended on \_\_\_\_\_

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with 37 C.F.R. 1.56. I hereby claim foreign priority benefits under 35 U.S.C. 119/365 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed or, if no priority is claimed, before the filing date of this application:

Priority Foreign Application(s):

Application Number	Country	Day/Month/Year Filed
JP 10-132654	Japan	April 27, 1998

I hereby claim the benefit under 35 U.S.C. §119(e) of any United States provisional application(s) listed below.

Application Number	Date/Month/Year Filed
--------------------	-----------------------

I hereby claim the benefit under 35 U.S.C. 120/365 of all prior United States and PCT international applications listed above or below and, insofar as the subject matter of each of the claims of this application is not disclosed in such prior applications in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose material information as defined in 37 C.F.R. 1.56 which occurred between the filing date of the prior applications and the national or PCT international filing date of this application:

Prior U.S./PCT Application(s):

Application Serial No.	Day/Month/Year Filed	Status: patented pending, abandoned
09/298,902	April 26, 1999	Abandoned

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon. And on behalf of the owner(s) hereof, I hereby appoint NIXON & VANDERHYE P.C., 1100 North Glebe Rd., 8<sup>th</sup> Floor, Arlington, VA 22201-4714, telephone number (703) 816-4000 (to whom all communications are to be directed), and the following attorneys thereof (of the same address) individually and collectively owner's/owners' attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith and with the resulting patent: Arthur R. Crawford, 25327; Larry S. Nixon, 25640; Robert A. Vanderhye, 27076; James T. Hosmer, 30184; Robert W. Faris, 31352; Richard G. Basha, 22770; Mark E. Nusbaum, 32348; Michael J. Keenan, 32106; Bryan H. Davidson, 30251; Stanley C. Spooner, 27393; Leonard C. Mitchard, 29009; Duane M. Byers, 33363; Jeffry H. Nelson, 30481; John R. Lastova, 33149; H. Warren Burnam, Jr. 29366; Thomas E. Byrne, 32205; Mary J. Wilson, 32955; J. Scott Davidson, 33489; Alan M. Kagen, 36178; Robert A. Molan, 29834; B. J. Sadoff, 36663; James D. Berquist, 34776; Updeep S. Gill, 37334; Michael J. Shea, 34725; Donald L. Jackson, 41090; Michelle N. Lester, 32331; Frank P. Presta, 19828; Joseph S. Presta, 35329; Joseph A. Rhoa, 37515; Raymond Y. Mah, 41426. I also authorize Nixon & Vanderhye to delete any attorney names/numbers no longer with the firm and to act and rely solely on instructions directly communicated from the person, assignee, attorney, firm, or other organization sending instructions to Nixon & Vanderhye on behalf of the owner(s).

1.	Inventor's Signature:	By: Yuko Takahashi, a Japanese citizen, and heiress-in-law of the estate of:	Date:
	Inventor:	Masayuki TAKAHASHI Japan (citizenship)	
	Residence: (city)	Tsurugashima-shi, Saitama MI (last) (state/country)	
	Post Office Address:	Reionzu-Mansion Wakaba Fujimi Kouen 13-1-301, Fujimi 2-chome, Tsurugashima-shi, Saitama, Japan	
	(Zip Code)	350-2201	
2.	Inventor's Signature:	Toshinori YUKINO Japan (citizenship)	Date: Aug. 2, 2002
	Inventor:	Toshinori YUKINO MI (last) (state/country)	
	Residence: (city)	Kitakatsushika-gun, Saitama Japan	
	Post Office Address:	Sakurada 3-5-1-7-203, Washimiya-machi, Kitakatsushika-gun, Saitama, Japan	
	(Zip Code)	340-0203	
3.	Inventor's Signature:	Mitsuru FUKUSHIMA Japan (citizenship)	Date: AUG 1, 2002
	Inventor:	Mitsuru FUKUSHIMA MI (last) (state/country)	
	Residence: (city)	Ohosato-gun, Saitama Japan	
	Post Office Address:	Youdo 2775, Yorii-machi, Ohosato-gun, Saitama, Japan	
	(Zip Code)	369-1201	

FOR ADDITIONAL INVENTORS, check box ☐ and attach sheet with same information and signature and date for each.

# *U.S. PATENT APPLICATION*

*Inventors:* Masayuki TAKAHASHI  
Toshinori YUKINO  
Mitsuru FUKUSHIMA

*Invention:* AUTOMOBILE INTERIOR OR EXTERIOR TRIM MATERIAL

NIXON & VANDERHYE P.C.  
ATTORNEYS AT LAW  
1100 NORTH GLEBE ROAD  
8<sup>TH</sup> FLOOR  
ARLINGTON, VIRGINIA 22201-4714  
(703) 816-4000  
Facsimile (703) 816-4100

## *SPECIFICATION*

Title: Automobile Interior or Exterior Trim Material

Cross-Reference to Related Application

This application is a continuation-in-part (CIP) of copending U.S. Patent Application Serial No. 09/298,902 filed on April 26, 1999, the entire content of which is expressly incorporated hereinto by reference.

## Automobile Interior or Exterior Trim Material

### BACKGROUND OF THE INVENTION

#### Field of the Invention

The present invention relates to material suitable for the interior or exterior trim of an automobile (hereinafter simply called an "interior or exterior trim material") which comprises a thermoplastic elastomer having improved weather resistance due to incorporation of a specific UV absorber; i.e., alkyl benzoate having a specified chemical structure. More particularly, the present invention relates to an interior or exterior trim material having improved weather resistance and resistance to weather-induced coloring.

#### Background Art

Conventionally, vinyl chloride resins have been widely used as interior or exterior trim materials. However, use of vinyl chloride resins has recently been restricted because of concerns in relation to environmental pollution, calling for substitution by non-halogen resins.

One possible substitute may be realized by use of a thermoplastic elastomer having resin properties suitable for an interior or exterior trim material. An interior or exterior trim material is required to have high weather resistance and resistance to weather-induced coloring, because the material is exposed to strong sunshine at high temperature while, for example, the automobile is parked outdoors.

Conventionally, a variety of UV absorbers and photostabilizers have been disclosed, and use method of these compounds has been suggested for imparting weather resistance to polyolefin resins used as polymer materials for general molding. Examples of UV absorbers include benzotriazole UV absorbers, benzophenone UV absorbers, phenoltriazine UV absorbers, and alkyl or aryl benzoate UV absorbers; and examples of photostabilizers include hindered amine photostabilizers.

For example, Japanese Patent Application Laid-Open (kokai) No. 54-21450 discloses a method for improving weather resistance of polyolefin resins by combined use of 3,5-di-tert-butyl-4-hydroxybenzoic acid hexadecyl ester (which is an alkyl benzoate UV absorber), and a benzotriazole UV absorber or a benzophenone UV absorber. Japanese Patent Application Laid-Open (kokai) No. 55-54339 discloses a method for improving weather resistance of polypropylene resins by combined use of the above-mentioned 3,5-di-tert-butyl-4-hydroxybenzoic acid hexadecyl ester and pentaerythritol dialkyldiphosphite. Japanese Patent Application Laid-Open (kokai) No. 56-62835 discloses a method for improving weather resistance of polyolefin by combined use of the above-mentioned 3,5-di-tert-butyl-4-hydroxybenzoic acid hexadecyl ester and a hindered amine photostabilizer.

Regarding the above-mentioned 3,5-di-tert-butyl-4-hydroxybenzoic acid hexadecyl ester, which is an alkyl benzoate UV absorber, Japanese Patent Application Laid-Open

(kokai) No. 58-84839 discloses use thereof for pipes made of poly-1-butene; Japanese Patent Application Laid-Open (kokai) No. 1-62360 discloses use of the same for polypropylene resins which are subjected to radiation sterilization and used in the field of medicine, and Japanese Patent Application Laid-Open (kokai) No. 7-188473 discloses use of the same as materials in the agricultural field.

Japanese Patent Application Laid-Open (kokai) No. 7-179719 discloses use of 3,5-di-tert-butyl-4-hydroxybenzoic acid 2,4-di-tert-butylphenyl ester (which is an aryl benzoate UV absorber) as an automobile material.

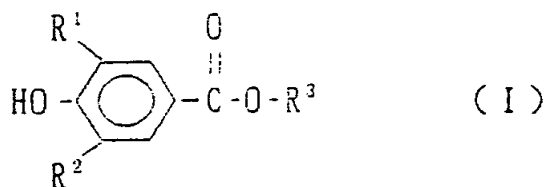
Thus, a variety of methods have been disclosed for improving weather resistance of polymer materials for general molding. However, no known UV absorbers or photostabilizers provide satisfactory effects. Thermoplastic elastomers which are to be used as substitutes for interior or exterior trim materials are no exception; they also involve the same problems. However, no method for improving properties of thermoplastic elastomers has been known. An object of the present invention is to solve the above problems inherent to thermoplastic elastomers.

#### SUMMARY OF THE INVENTION

In view of the foregoing, the present inventors have conducted careful studies and have found that, when a 3,5-dialkyl-4-hydroxybenzoic acid alkyl ester is added to a thermoplastic elastomer, there can be obtained an interior or

exterior trim material which has excellent weather resistance, and resistance to weather-induced coloring, without causing bleeding or fogging, or contamination, such as plate-out, of processing apparatuses. The 3,5-dialkyl-4-hydroxybenzoic acid alkyl ester is an UV absorber of alkyl benzoate among other benzoate compounds.

Accordingly, in a first aspect of the present invention, there is provided an interior or exterior trim material which comprises a thermoplastic elastomer composition containing 100 parts by weight of a thermoplastic elastomer and 0.001-10 parts by weight of an alkyl benzoate compound represented by the following formula (I):



wherein each of  $\text{R}^1$  and  $\text{R}^2$  is a hydrogen atom, a C1-C8 alkyl or cycloalkyl group, or a C6-C12 aryl, alkylaryl, or arylalkyl group; and  $\text{R}^3$  is a C1-C30 alkyl group.

Preferably, each of  $\text{R}^1$  and  $\text{R}^2$  in the alkyl benzoate compound represented by formula (I) is a C1-C8 alkyl group.

Preferably, each of  $\text{R}^1$  and  $\text{R}^2$  in the alkyl benzoate compound represented by formula (I) is a tertiary butyl group or a tertiary amyl group.

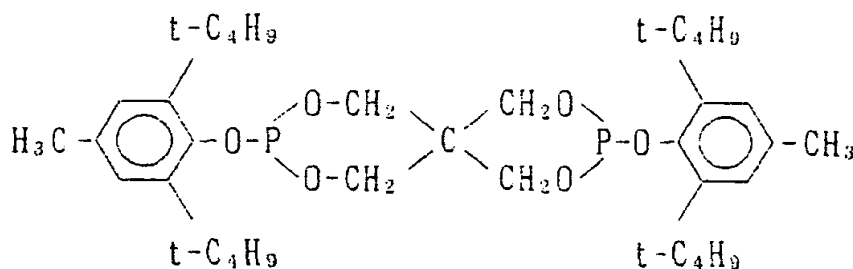
Preferably,  $\text{R}^3$  in the alkyl benzoate compound represented by formula (I) is a C6-C8 alkyl group.

Preferably, the thermoplastic elastomer is a polyolefin thermoplastic elastomer.

Preferably, the interior or exterior trim material further contains a hindered amine photostabilizer.

Preferably, the hindered amine photostabilizer has a 1,2,2,6,6-pentamethyl-4-piperidyl group.

Preferably, the interior or exterior trim material further contains a phosphorus-containing antioxidant of the following formula in an amount of 0.001-10 parts by weight.



Preferably, the interior or exterior trim material further contains a pigment.

Preferably, the interior or exterior trim material is used as a facing material for a ceiling, seat, or dashboard.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

##### Thermoplastic elastomers

Examples of thermoplastic elastomers used in the present invention include polyolefin thermoplastic elastomers and block copolymer-type polystyrene thermoplastic elastomers. The polyolefin thermoplastic elastomer comprises polyolefin resins such as polypropylene and polyethylene serving as hard

segments and rubber compositions such as EPDM serving as soft segments. The block copolymer-type polystyrene thermoplastic elastomer comprises polystyrene serving as hard segments and polydienes such as polybutadiene or polyisoprene serving as soft segments. Alternatively, a blend of the polyolefin elastomers and the polystyrene elastomers may also be used as the thermoplastic elastomer of the present invention.

The methods for combining soft segments and hard segments in thermoplastic elastomers may be roughly divided into simple blending, implantation by copolymerization, and dynamic cross-linking.

Combinations of segments of polystyrene thermoplastic elastomers include a styrene-butadiene-styrene block copolymer (SBS), a styrene-isoprene-styrene block copolymer (SIS), a styrene-ethylene-butylene-styrene block copolymer (SEBS), a styrene-ethylene-propylene-styrene block copolymer (SEPS), a hydrogenated polymer of any one of the four copolymers, a hydrogenated polymer of random SBR (HSBR), and a blend of polypropylene and one or more arbitrary members selected from among these polymers.

Alkyl benzoate compounds represented by formula (I)

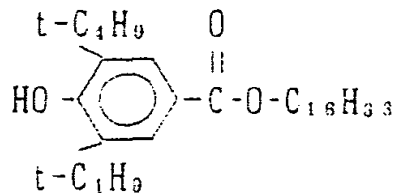
Examples of alkyl groups represented by  $R^1$  and  $R^2$  in the above formula (I) include alkyl groups such as methyl, ethyl, propyl, isopropyl, butyl, isobutyl, sec-butyl, tert-butyl, pentyl, tert-pentyl, hexyl, heptyl, octyl, and tert-octyl; cycloalkyl groups such as cyclopentyl and cyclohexyl; aryl groups such as phenyl and naphthyl; alkylaryl groups such as

methylphenyl and butylphenyl; and arylalkyl groups such as phenylmethyl, 1-phenylethyl, and cumyl.

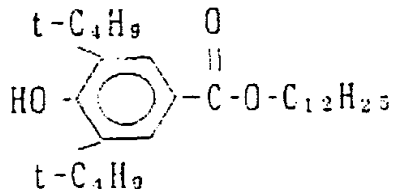
Examples of alkyl groups represented by  $R^1$  include methyl, ethyl, propyl, isopropyl, butyl, isobutyl, secondary-butyl, tertiary-butyl, pentyl, tertiary-pentyl, hexyl, heptyl, octyl, tertiary-octyl, 2-ethylhexyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, octadecyl, behenyl, and triacontyl.

More specifically, examples of the alkyl benzoate compounds represented by formula (I) include the following compounds (Nos. 1-5). However, the present invention is in no way limited by the following illustrations.

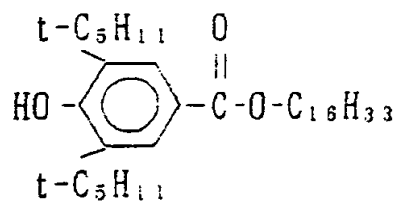
Compound No. 1:



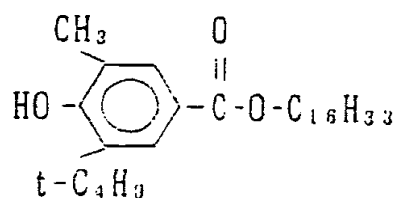
Compound No. 2:



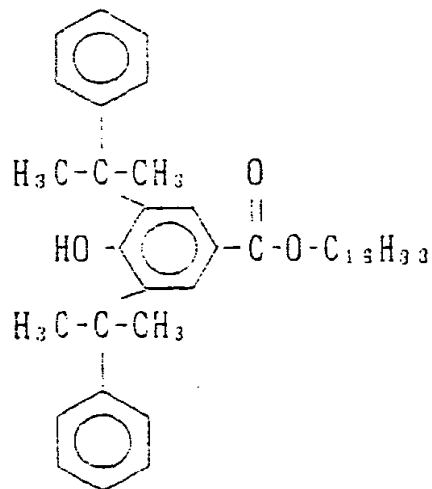
Compound No. 3:



Compound No. 4:



Compound No. 5:



The thermoplastic elastomer composition of the present invention comprises the aforementioned thermoplastic elastomer (100 parts by weight) and the aforementioned alkyl benzoate compound (0.001-10 parts by weight). When the amount of the alkyl benzoate compound is less than 0.001 parts by weight, sufficient weather resistance required for

an interior or exterior trim material cannot be obtained. In contrast, an amount thereof in excess of 10 parts by weight is not preferable, because the compound may bleed from the interior or exterior trim material because of a change in the environment, such as a rise in temperature.

The thermoplastic elastomer composition of the present invention is appropriately used as interior or exterior trim materials; for example, for materials of ceilings, doors, seats, trunks, wipers and bumper, which are formed by use of known methods such as extrusion molding, injection molding, compression molding, or lamination. The thermoplastic elastomers are preferably used as interior trim materials for ceilings, seats, and dashboards.

Hindered amine photo-stabilizers may optionally be added into the thermoplastic elastomer composition of the present invention. The addition of the hindered amine photo-stabilizers advantageously amplifies the effect of aforementioned alkyl benzoate compounds used in the present invention; specifically, enhancement in weather resistance.

In addition, additives such as widely-used antioxidants may be used in combination with the above composition, and examples of the antioxidants include a phosphite compound, a phenol compound, and a sulfur compound.

Ultraviolet absorbers other than the alkyl benzoate compounds of the present invention may be used in combination with the above-described composition.

Examples of the above-described hindered amine photo-

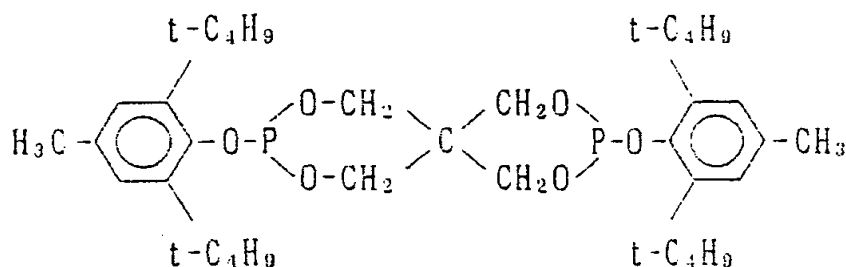
stabilizers include 2,2,6,6-tetramethyl-4-piperidyl benzoate,  
 N-(2,2,6,6-tetramethyl-4-piperidyl)dodecylsuccinic imide, 1-  
 [(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxyethyl]-  
 2,2,6,6-tetramethyl-4-piperidyl(3,5-di-tert-butyl-4-  
 hydroxyphenyl)propionate, bis(2,2,6,6-tetramethyl-4-  
 piperidyl) sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)  
 sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)-2-butyl-2-  
 (3,5-di-tert-butyl-4-hydroxybenzyl) malonate, N,N'-  
 bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine,  
 tetra(2,2,6,6-tetramethyl-4-piperidyl)  
 butanetetracarboxylate, bis(2,2,6,6-tetramethyl-4-  
 piperidyl)'di(tridecyl) butanetetracarboxylate,  
 bis(1,2,2,6,6-pentamethyl-4-piperidyl)'di(tridecyl)  
 butanetetracarboxylate, 3,9-bis[1,1-dimethyl-2-{tris(2,2,6,6-  
 tetramethyl-4-piperidyloxycarbonyloxy)butylcarbonyloxy}-  
 ethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, 3,9-bis[1,1-  
 dimethyl-2-{tris(1,2,2,6,6-pentamethyl-4-  
 piperidyloxycarbonyloxy)butylcarbonyloxy}ethyl]-2,4,8,10-  
 tetraoxaspiro[5.5]undecane, 1,5,8,12-tetrakis[4,6-bis{N-  
 (2,2,6,6-tetramethyl-4-piperidyl)butylamino}-1,3,5-triazin-2-  
 yl]-1,5,8,12-tetrazadodecane, a 1-(2-hydroxyethyl)-2,2,6,6-  
 tetramethyl-4-piperidinol/dimethyl succinate condensation  
 product, a 2-tert-octylamino-4,6-dichloro-s-triazine/N,N'-  
 bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine  
 condensation product, an N,N'-bis(2,2,6,6-tetramethyl-4-  
 piperidyl)hexamethylenediamine/dibromoethane condensation  
 product, 2,2,6,6-tetramethyl-4-hydroxypiperidin-N-oxyl,

bis(2,2,6,6-tetramethyl-N-oxypiperidine) sebacate, tetrakis(2,2,6,6-tetramethyl-N-oxypiperidyl) butane-1,2,3,4-tetracarboxylate, tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl) 1,2,3,4-butanetetracarboxylate, and 3,9-bis(1,1-dimethyl-2-(tris (2, 2, 6, 6-tetramethyl-N-oxypiperidyl-4-oxycarbonyl)butylcarbonyloxy) ethyl) -2,4,8, 10-tetraoxaspiro [5.5] undecane, especially carboxylic acid esters of 1,2,2,6,6-pentamethyl-4-piperidinol or 2,2,6,6-tetramethyl-4-piperidinol.

The hindered amine photostabilizer is most preferably present in an amount between about 0.10 to about 0.50 part by weight, for example, between about 0.15 to about 0.30 part by weight, based on 100 parts by weight of the thermoplastic elastomer.

Examples of the phosphite antioxidants include tris(nonylphenyl) phosphite, tris(2,4-di-tert-butylphenyl) phosphite, tris[2-tert-butyl-4- (3-tert-butyl-4-hydroxy-5-methylphenylthio) -5-methylphenyl] phosphite, tridecyl phosphite, octyldiphenyl phosphite, di (decyl)monophenyl phosphite, di (tridecyl) pentaerythritol diphosphite, distearyl pentaerythritol diphosphite, di (nonylphenyl)pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tri-tert-butylphenyl)pentaerythritol diphosphite, bis (2,4-dicumylphenyl)pentaerythritol diphosphite, tetra(tridecyl) isopropylidenediphenol diphosphite, tetra(tridecyl)-4,4'-n-butylidenebis(2-tert-butyl-5-methylphenol) diphosphite, hexa(tridecyl) -1,1,3-tris(2-methyl-4-hydroxy-5-tert-butylphenyl)butane triphosphite, 2,2'-methylenebis(4,6-di-tert-butylphenyl) -2-ethylhexyl phosphite, 2,2'-methylenebis(4,6-di-tert-butylphenyl)octadecyl phosphite, 2,2'-ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite, tetrakis(2,4-di-tert-

butylphenyl)biphenylene diphosphonite, 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide, tris(2-[(2,4,8,10-tetrakis(tert-butyl)dibenzo[d,f][1,3,2]dioxaphosphebine-6-yl)oxy]ethyl)amine, and a phosphorous acid ester of 2-ethyl-2-butylpropylene glycol and 2,4,6-tri-tert-butyl phenol. Of these, bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite represented by the following formula is preferable in that it provides particularly excellent stabilizing effect.



Examples of the phenol antioxidants include 2,6-di-tert-butyl-p-cresol, 2,6-diphenyl-4-octadecyloxyphenol, stearyl (3,5-di-tert-butyl-4-hydroxyphenyl) propionate, distearyl (3,5-di-tert-butyl-4-hydroxybenzyl)phosphonate, thiodiethylene glycol bis[(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], 1,6-hexamethylenebis[(3,5-di-tert-butyl-4-hydroxyphenyl)propionate], 1,6-hexamethylenebis[(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid amide], 4,4'-thiobis(6-tert-butyl-m-cresol), 2,2'-methylenebis(4-methyl-6-tert-butylphenol), 2,2'-methylenebis(4-ethyl-6-tert-butylphenol), a bis[3,3-bis(4-hydroxy-3-tert-butylphenyl)butyric acid] glycol ester, 4,4'-butylidenebis(6-

tert-butyl-m-cresol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4-sec-butyl-6-tert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-5-tert-butylphenyl)butane, bis[2-tert-butyl-4-methyl-6-(2-hydroxy-3-tert-butyl-5-methylbenzyl)phenyl] terephthalate, 1,3,5-tris(2,6-dimethyl-3-hydroxy-4-tert-butylbenzyl) isocyanurate, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl) isocyanurate, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,3,5-tris[(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxyethyl] isocyanurate, tetrakis[methylene-3-(3',5'-di-tert-butyl-4'-hydroxyphenyl)propionate]methane, 2-tert-butyl-4-methyl-6-(2-acryloyloxy-3-tert-butyl-5-methylbenzyl)phenol, 3,9-bis[1,1-dimethyl-2-[(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy]ethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, and triethylene glycol bis[(3-tert-butyl-4-hydroxy-5-methylphenyl)propionate].

Examples of the sulfur-containing antioxidants include dialkyl thiodipropionates such as dilauryl thiodipropionate, dimyristyl thiodipropionate, and distearyl thiodipropionate; and  $\beta$ -alkylmercaptopropionic acid esters of polyol such as pentaerythritol tetra( $\beta$ -dodecylmercaptopropionate).

Examples of the ultraviolet absorbers other than the alkylbenzoate compounds of the present invention include 2-hydroxybenzophenones such as 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-octoxybenzophenone, and 5,5'-methylenebis(2-hydroxy-4-methoxybenzophenone); 2-(2'-hydroxyphenyl)benzotriazols such as 2-(2'-hydroxy-5'-

methylphenyl)benzotriazol, 2-(2'-hydroxy-3',5' i-tert-butylphenyl)benzotriazol, 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)-5-chlorobenzotriazol, 2-(2'-hydroxy-3'-tert-butyl-5'-methylphenyl)-5-chlorobenzotriazol, 2-(2'-hydroxy-5'-tert-octylphenyl)benzotriazol, 2-(2'-hydroxy-3',5'-dicumylphenyl)benzotriazol, and 2,2'-methylenebis(4-tert-octyl-6-benzotriazolyl)phenol; benzoates such as phenyl salicylate, resorcinol monobenzoate, and 2,4-di-tert-butylphenyl 3',5'-di-tert-butyl-4'-hydroxybenzoate; substituted oxanilides such as 2-ethyl-2'-ethoxyoxanilide and 2-ethoxy-4'-dodecyloxanilide; and cyanoacrylates such as ethyl  $\alpha$ -cyano- $\beta,\beta$ -diphenylacrylate and methyl 2-cyano-3-methyl-3-(p-methoxyphenyl)acrylate.

The thermoplastic elastomer composition of the present invention may also comprise heavy metal deactivators, nucleating agents, metallic soap, hydrotalcites, pigments, organic tin compounds, plasticizers, epoxy compounds, foaming agents, antistatic agents, flame retardants, lubricants, processing aids, and inorganic fillers, such as talc, silica, mica, calcium silicate, calcium carbonate, barium sulfate, zinc oxide, magnesium hydroxide or a mixture thereof. The inorganic filler is incorporated in an amount of not more than 30 parts by weight, based on 100 parts by weight of the thermoplastic elastomer.

Particularly, in the thermoplastic elastomer composition of the present invention, the color of the product is easily adjusted, and various known pigments can be used for the product, since plate-out of pigments from the composition into a processing machine does not occur.

Examples of pigments used in the thermoplastic elastomer composition of the present invention include inorganic pigments, azo pigments, nitro pigments, acine

pigments, acidic dye lake pigments, vat dye pigments, isoindolinone pigments, basic dye lake pigments, mordant dye pigments, quinacridone pigments, phthalocyanine pigments, nitroso pigments, daylight fluorescent pigments, metal powder pigments, and polymer coupled pigments.

When the thermoplastic elastomers of the present invention are produced, the methods for addition of alkylbenzoate compounds and other additives thereto are not limited, and additives may be used in the following forms: powder, water dispersions such as emulsions or suspensions, and organic solutions.

Although the types of blenders are not limited, when powder additives are used, a ribbon mixer or a Henschel mixer is useful for dry blending, and a uniaxial or biaxial extruder is useful for kneading. A conventional vertical mixer is sufficient for blending when additives assume the forms of water dispersions or solutions.

Additionally, various processes for addition may be employed in accordance with the forms of additives. For example, powder additives can be added during molding of the thermoplastic elastomer compositions, and adsorption or impregnation of additives into a molded product may be performed by dipping a molded product into an additive solution after molding. As another method for addition during molding, the additives may be kneaded in a final blending process after preparation of a blended master batch of high concentration, or through granulation of powder

additives to depress a dust.

The present invention will next be described in more detail by way of examples, which should not be construed as limiting the invention thereto.

Example 1

Propylene resin (60 parts by weight), EPR (20 parts by weight), talc (20 parts by weight), titanium oxide (2 parts by weight), calcium stearate (0.05 parts by weight), stearyl (3,5-di-tert-butyl-4-hydroxyphenyl)propionate (0.1 parts by weight), tris(2,4-di-tert-butylphenyl) phosphite (0.05 parts by weight), tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl) 1,2,3,4-butanetetracarboxylate (0.15 parts by weight) and an alkyl benzoate compound (Compound No. 1) (0.1 parts by weight) were blended in a ribbon mixer. The resultant mixture was supplied to a pelletizer and extruded at 250°C, to thereby prepare pellets.

Subsequently, after the above pellets were melted at 250°C, 2-mm thick sheets were prepared by injection molding, so as to serve as an interior or exterior trim material. The sheets were tested for weather resistance (time until occurrence of cracking) and weather coloring (yellowness index after 480 hours) by use of a sunshine weatherometer under the following conditions: black panel temperature: 83°C, 18-minute rain in a 120-minute cycle.

Ten test pieces (25 mm × 50 mm × 2 mm) of an interior or exterior trim material were placed in an 80°C oven, and after elapse of one week their surfaces were visually observed for

occurrence of bloom, as well as for severity of any bloom found. The results are evaluated as follows. O: no bloom, X: bloom on the entire surface.

A test tube containing 25 grams of a specimen was covered with a glass plate, and heated in a 100°C oil bath for 48 hours, to thereby check for presence of deposited matter and the degree thereof (fogging) in the same manner as the bloom test. The results are evaluated as follows. O: no deposition over the glass plate, X: deposition over the entire glass surface. The results are shown in Table 1.

#### Comparative Example 1

The same procedure as in Example 1 was performed, except that no alkyl benzoate compound was used (Comparative Example 1-1), an aryl benzoate compound serving as a UV absorber was used in place of the above compound (Comparative Example 1-2), or a UV absorbing compound other than a benzoate compound was used in place of the above compound (Comparative Example 1-3). The results are shown in Table 1.

Table 1

	UV absorber	Time at which cracking occurred (hr)	Yellowness Index	Bloom	Fogging
Example 1	Compound No. 1	2900	2.3	O	O
Comp Ex. 1-1	None	1000	5.1	O	O
Comp Ex. 1-2	Comp. Compound 1 <sup>*</sup>	2100	12.2	X	X
Comp Ex. 1-3	Comp. Compound 2 <sup>**</sup>	1100	4.5	X	X

\*<sup>1</sup>: 3,5-di-tert-butyl-4-hydroxybenzoic acid-2,4-di-tert-butylphenyl ester

\*<sup>2</sup>: 2-(2'-hydroxy-5'-methylphenyl)-5-chlorobenzotriazole

### Example 2

The procedure of Example 1 was performed, except that propylene resin (65 parts by weight), EPDM (15 parts by weight), talc (20 parts by weight), titanium oxide (2 parts by weight), Phthalocyanine Blue (1 part by weight), hydrotalcite (0.1 parts by weight), tetrakis[methylene-3-(3',5'-di-tert-butyl-4'-hydroxyphenyl)propionate]methane (0.1 parts by weight), bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite (0.1 parts by weight), 3,9-bis[1,1-dimethyl-2-{tris(1,2,2,6,6-pentamethyl-4-piperidyloxycarbonyloxy)butylcarbonyloxy}ethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane (0.2 parts by weight), and alkylbenzoate (Compound No. 1) (0.3 parts by weight) were blended, to thereby prepare pellets. By use of a kneading roller at 240°C, the pellets were molded into a sheet having a thickness of 0.1 mm.

A test piece of 25 mm × 50 mm × 0.1 mm was prepared from the obtained sheet, and the time at which cracking of the piece initiated was measured under the same conditions as in Example 1, by use of a sunshine weatherometer, to thereby evaluate the weather resistance thereof. Bloom and fogging of the piece were also evaluated in the same manner as in Example 1. The results of evaluation are shown in Table 2.

### Comparative Example 2

The procedure of Example 2 was performed, except that no alkylbenzoate was used (Comparative Example 2-1), arylbenzoate was used as an ultraviolet absorber (Comparative

Example 2-2), or a compound other than a benzoate compound was used as an ultraviolet absorber (Comparative Example 2-3). The results are shown in Table 2.

Table 2

	UV absorbers	Time at which cracking occurred (hr)	Bloom	Fogging
Example 2	Compound No. 1	2700	O	O
Comp. Ex.				
2-1	None	800	O	O
2-2	Comp. Compound 1 <sup>*1</sup>	1000	X	X
2-3	Comp. Compound 2 <sup>*2</sup>	1800	X	X

\*1 and \*2 correspond to the compounds of the same number shown in Table 1.

### Example 3

The procedure of Example 2 was performed, except that propylene-ethylene copolymer (70 parts by weight), SBS (30 parts by weight), Phthalocyanine Blue (1 part by weight), calcium stearate (0.2 parts by weight), tetrakis[methylene-3-(3',5'-di-tert-butyl-4'-hydroxyphenyl)propionate]methane (0.2 parts by weight), bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite (0.2 parts by weight), bis(2,2,6,6-tetramethyl-4-piperidyl) sebacate (0.3 parts by weight), and alkylbenzoate compound (Compound No. 1) (0.3 parts by weight) were blended, to thereby prepare pellets. By use of a kneading roller at 240°C, the pellets were molded into a sheet having a thickness of 0.1 mm.

A test piece was prepared from the sheet in the same

manner as in Example 2, and weather resistance and resistance to weather-induced coloring thereof were evaluated by use of a sunshine weatherometer in the same manner as in Example 1 except that rainfall cycling was omitted. Weather resistance was evaluated in terms of degradation time, and resistance to weather-induced coloring was evaluated in terms of yellowness index after 2000 hours.

Transfer of pigments from the piece to the kneading roller resulting from the additives was considered to constitute plate-out. Specifically, after preparation of the aforementioned compounds, a sheet was prepared, by use of a kneading roller, from a vinyl chloride resin containing a white pigment. Blue-coloring of the sheet of vinyl chloride resin was evaluated by visual inspection as follows, X: obvious coloring,  $\Delta$ : slight coloring, 0: no coloring. The results are shown in Table 3.

#### Comparative Example 3

The procedure of Example 3 was performed, except that arylbenzoate was used as an ultraviolet absorber in place of the alkylbenzoate compound (Comparative Example 3-1), or a compound other than a benzoate compound was used as an ultraviolet absorber (Comparative Example 3-2). The results are shown in Table 3.

Table 3

	Test Compound	Time until embrittlement (hr) (weather resistance)	Plate- out	Yellowing (resistance to weather- induced coloring)
Example 3	Compound No. 1	3300	O	4.6
Comp. Ex. 3-1	Comp. Compound 1 <sup>*1</sup>	2300	Δ	15.6
Comp. Ex. 3-2	Comp. Compound 2 <sup>*2</sup>	1500	X	10.8

\*1 and 2\* correspond to the compounds of the same number shown in Table 1.

#### Example 4

Propylene resin (60 parts by weight), EPR (20 parts by weight), talc (20 parts by weight), titanium oxide (2 parts by weight), calcium stearate (0.05 parts by weight), tetrakis[methylene-3-(3',5'-di-tert-butyl-4'-hydroxyphenyl)propionate]methane (0.1 parts by weight), tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl) 1,2,3,4-butanetetracarboxylate (0.15 parts by weight), alkylbenzoate compound (Compound No. 1) (0.2 parts by weight), and additives shown in Table 4 (0.1 parts by weight) were blended, and the resultant mixture was extruded at 250°C to prepare pellets. Subsequently, a sheet having a thickness of 2 mm was prepared from the pellets through injection molding at 250°C, and the yellowness index thereof was measured. Weather resistance and resistance to weather-induced coloring were evaluated by use of a sunshine weatherometer in the same manner as in Example 1, except that rainfall cycling was omitted. Weather resistance was evaluated in terms of time

until initiation of cracking, and resistance to weather-induced coloring was evaluated in terms of yellowness index. The results are shown in Table 4.

Table 4

	Additives	Time until initiation of cracking (hr)	Yellowness Index
Example 4-1	none	1400	10.2
4-2	2112 <sup>*3</sup>	1400	6.2
4-3	HP-10 <sup>*4</sup>	1600	5.1
4-4	PEP-36 <sup>*5</sup>	2200	3.0

\*3: tris(2,4-di-tert-butylphenyl) phosphite

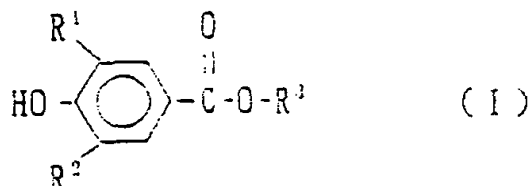
\*4: 2,2'-methylenebis(4,6-di-tert-butylphenyl)-2-ethylhexyl phosphite

\*5: bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite

As clearly seen in the above mentioned detailed description, especially the results of Examples, when improving weather resistance of a thermoplastic elastomer resin, a thermoplastic elastomer containing the alkylbenzoate compound according to the present invention as an ultraviolet absorber exhibits remarkably improved weather resistance and resistance to weather-induced coloring as compared with the case where antioxidants, hindered amine photo-stabilizer, or other ultraviolet absorbers are used. Use of the alkylbenzoate compound yields other improvements, including prevention of blooming, prevention of fogging, prevention of plate-out during processing, and prevention of staining of a processing machine, because pigments which are used are not transferred.

What is Claimed is:

1. An automobile interior or exterior trim material which comprises a thermoplastic elastomer composition containing 100 parts by weight of a thermoplastic elastomer; 0.001-10 parts by weight of an alkyl benzoate compound represented by the following formula (I):



wherein each of  $\text{R}^1$  and  $\text{R}^2$  is a hydrogen atom, a C1-C8 alkyl or cycloalkyl group, or a C6-C12 aryl, alkylaryl, or arylalkyl group; and  $\text{R}^3$  is a C1 - C30 alkyl group; and a hindered amine photostabilizer.

2. An automobile interior or exterior trim material according to Claim 1, wherein each of  $\text{R}^1$  and  $\text{R}^2$  in the alkyl benzoate compound represented by formula (I) is a C1-C8 alkyl group.

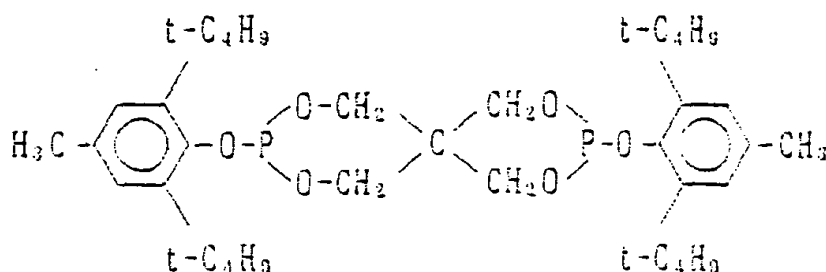
3. An automobile interior or exterior trim material according to Claim 1, wherein each of  $\text{R}^1$  and  $\text{R}^2$  in the alkyl benzoate compound represented by formula (I) is a tertiary butyl group or a tertiary amyl group.

4. An automobile interior or exterior trim material according to Claim 1, wherein  $\text{R}^1$  in the alkyl benzoate compound represented by formula (I) is a C6-C8 alkyl group.

5. An automobile interior or exterior trim material according to Claim 1, wherein the thermoplastic elastomer is a polyolefin thermoplastic elastomer.

6. An automobile interior or exterior trim material according to Claim 1, wherein the hindered amine photostabilizer has a 1,2,2,6,6-pentamethyl-4-piperidyl group.

7. An automobile interior or exterior trim material according to Claim 1, which further contains a phosphorus-containing antioxidant of the following formula in an amount of 0.001-10 parts by weight:



8. An automobile interior or exterior trim material according to Claim 1, which further contains a pigment.

9. An automobile interior or exterior trim material according to Claim 1, which is used as a facing material for a ceiling, seat, or dashboard.

10. An automobile interior or exterior trim material according to Claim 1, 2 or 3, wherein  $R^3$  is a C12-C16 alkyl group.

11. An automobile interior or exterior trim material according to claim 1, wherein the hindered amine photostabilizer is present in an amount between 0.10 to 0.50 part by weight, based on 100 parts by weight of the thermoplastic elastomer.

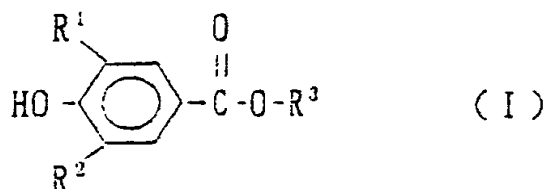
12. An automobile interior or exterior trim material according to Claim 1, wherein the hindered amine photostabilizer is present in an amount between 0.15 to 0.30 part by weight, based on 100 parts by weight of the thermoplastic elastomer.

13. An automobile interior or exterior trim material according to claim 1, wherein the hindered amine photostabilizer is a carboxylic acid ester of 1,2,2,6,6-pentamethyl-4-piperidinol or 2,2,6,6-tetramethyl-4-piperidinol.

14. An automobile interior or exterior trim material according to claim 1, wherein the hindered amine photostabilizer is selected from among tetrakis (1,2,2,6,6-pentamethyl-4-piperidyl) 1,2,3,4-butanetetracarboxylate, 3,9-[1,1-dimethyl-2-{tris(1,2,2,6,6-pentamethyl-4-piperidyloxycarbonyloxy) butylcarbonyloxy} ethyl] - 2,4,8,10-tetraoxaspiro [5.5] undecane, and bis (2,2,6,6-tetramethyl-4-piperidyl) sebacate.

# ABSTRACT

An automobile interior or exterior trim material which is formed of a thermoplastic elastomer composition containing 100 parts by weight of a thermoplastic elastomer and 0.001-10 parts by weight of an alkyl benzoate compound of the following formula (I):



wherein each of  $\text{R}^1$  and  $\text{R}^2$  is a hydrogen atom, a C1-C8 alkyl or cycloalkyl group, or a C6-C12 aryl, alkylaryl, or arylalkyl group; and  $\text{R}^3$  is a C1-C30 alkyl group. The interior or exterior trim material of the invention has improved weather resistance and resistance to weather-induced coloring.

(19)日本国特許庁 (J P)

(12) 公 開 特 許 公 報 (A)

(11)特許出願公開番号

特開平11-310667

(43)公開日 平成11年(1999)11月9日

(51)Int.Cl.<sup>6</sup>

識別記号

F I

C 0 8 L 21/00

C 0 8 L 21/00

B 2 9 C 47/00

B 2 9 C 47/00

C 0 8 K 5/101

C 0 8 K 5/101

5/34

5/34

5/524

5/524

審査請求 未請求 請求項の数 4 F D (全 9 頁) 最終頁に続く

(21)出願番号

特願平10-132654

(22)出願日

平成10年(1998)4月27日

(71)出願人 000000387

旭電化工業株式会社

東京都荒川区東尾久7丁目2番35号

(72)発明者 高橋 雅之

埼玉県浦和市白幡5丁目2番13号 旭電化  
工業株式会社内

(72)発明者 幸野 俊則

埼玉県浦和市白幡5丁目2番13号 旭電化  
工業株式会社内

(72)発明者 福島 充

埼玉県浦和市白幡5丁目2番13号 旭電化  
工業株式会社内

(74)代理人 弁理士 三浦 良和

(54)【発明の名称】 自動車内外装材

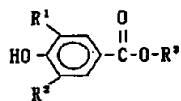
(57)【要約】

【課題】自動車内外装材に使用される熱可塑性エラストマーについて、その耐候性、耐光着色性等を改善する手段を提供すること。

【解決手段】熱可塑性エラストマー100重量部に、一

般式(Ⅰ)で表されるアルキルベンゾエート化合物0.001~10重量部が添加された熱可塑性エラストマー組成物からなる自動車内外装材。

【化1】

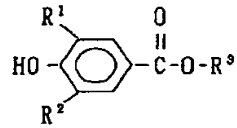


(1)

(式中、R<sup>1</sup> および R<sup>2</sup> は各々独立に水素原子、炭素原子数1~8のアルキル基またはシクロアルキル基、または炭素原子数6~12のアリール基、アルキルアリール基またはアリールアルキル基を表し、R<sup>3</sup>は炭素原子数1~30のアルキル基を表す。)

## 【特許請求の範囲】

【請求項1】 熱可塑性エラストマー100重量部に、下記一般式(I)で表されるアルキルベンゾエート化合



物0.001~10重量部が添加された熱可塑性エラストマー組成物からなる自動車内外装材。

【化1】

(I)

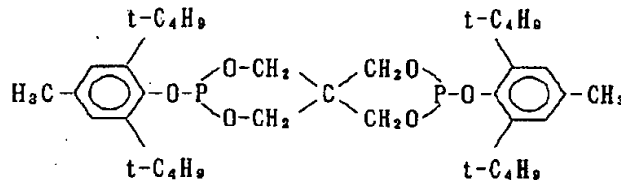
(式中、R¹ およびR² は各々独立に水素原子、炭素原子数1~8のアルキル基またはシクロアルキル基、または炭素原子数6~12のアリール基、アルキルアリール基またはアリールアルキル基を表し、R³は炭素原子数1~30のアルキル基を表す。)

【請求項2】 熱可塑性エラストマーが、ポリオレフィン系熱可塑性エラストマーである請求項1記載の自動車内外装材。

【請求項3】 ヒンダードアミン系光安定剤が添加された請求項2記載の自動車内外装材。

【請求項4】 下記構造式を有するリン系酸化防止剤0.001~10重量部が添加された請求項3記載の自動車内外装材。

【化2】



## 【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、特定の構造を有するアルキルベンゾエート系化合物の紫外線吸収剤により耐候性が改善された熱可塑性エラストマーからなる自動車内外装材に関する。より詳細には、耐候性および耐光着色性に優れた自動車内外装材に関する。

【0002】

【従来の技術】自動車内外装材としては、従来塩化ビニル樹脂が広く用いられてきた。しかし、塩化ビニル樹脂は環境汚染の懸念から使用が制限されるようになり、非ハロゲン系樹脂による代替が求められている。

【0003】そこで、自動車内外装材に適した樹脂物性を有する熱可塑性エラストマーによる代替が検討されている。しかし、自動車内外装材は屋外での駐車などに際して高温下で強い日差しに曝されるため、高度の耐候性、耐候着色性が要求されている。

【0004】従来、一般成形用の高分子材料としてのポリオレフィン系樹脂への耐候性付与の目的で、種々の紫外線吸収剤や光安定剤が開示され、又使用方法が提案されてきた。前者の紫外線吸収剤としては例えばベンゾトリアゾール系紫外線吸収剤、ベンゾフェノン系紫外線吸収剤、フェノールトリアジン系紫外線吸収剤、アルキル又はアリールベンゾエート系紫外線吸収剤等が開示さ

れ、後者の光安定剤としてはヒンダードアミン系光安定剤等が開示されている。

【0005】例えば特開昭54-21450号公報には、ポリオレフィン系樹脂の耐候性改善剤方法としてアルキルベンゾエート系紫外線吸収剤に属する3,5-ジ第三ブチル-4-ヒドロキシ安息香酸ヘキサデシルエステルとベンゾトリアゾール系紫外線吸収剤またはベンゾフェノン系紫外線吸収剤の併用方法が、特開昭55-54339号公報には、ポリプロピレン系樹脂の耐候性改善剤方法として上記3,5-ジ第三ブチル-4-ヒドロキシ安息香酸ヘキサデシルエステルとベンタエリスリトールジアルキルジホスファイトの併用方法が、また特開昭56-62835号公報には、上記3,5-ジ第三ブチル-4-ヒドロキシ安息香酸ヘキサデシルエステルとヒンダードアミン系光安定剤の併用がそれぞれ提案されている。

【0006】更に、上記アルキルベンゾエート系紫外線吸収剤に属する3,5-ジ第三ブチル-4-ヒドロキシ安息香酸ヘキサデシルエステルは、特開昭58-84839号公報にはポリ-1-ブテンからなるパイプに用いられることが、特開平1-62360号公報には放射線滅菌が施される医療用ポリプロピレンに用いられることが、また特開平7-188473号公報には、農業用資材に用いられることがそれぞれ提案されている。一方、

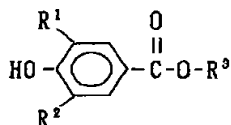
特開平7-179719号公報には、アリアルベンゾエート系紫外線吸収剤に属する3,5-ジ第三ブチル-4-ヒドロキシ安息香酸-2,4-ジ第三ブチルフェニルエステルを自動車材料に用いることが提案されている。

【0007】

【発明が解決しようとする課題】このように一般成形用の高分子材料には各種耐候性改善方法が提案されているが、各種公知紫外線吸収剤、光安定剤等もその効果においては充分満足のいくものではない。自動車用内外装材として代替使用されようとしている熱可塑性エラストマーについても同様の問題点があるにもかかわらず、なんらその改善方法が知られていないのが現状であり、本発明は熱可塑性エラストマーについてのこの点を解決することを課題とするものである。

【0008】

【課題を解決するための手段】本発明者等は上記の現状



に鑑み、鋭意検討を重ねた結果、ベンゾエート系紫外線吸収剤のうちアルキルベンゾエート系化合物の紫外線吸収剤に相当する3,5-ジアルキル-4-ヒドロキシ安息香酸アルキルエステルが添加された熱可塑性エラストマーは、プレートアウトなどの加工機汚染、ブリードおよびフォギングの問題なしに、耐光着色性等の耐候性に優れた自動車内外装材を提供できることを見出し、本発明に到達した。

【0009】即ち、本発明は熱可塑性エラストマー100重量部に、下記一般式(I)で表されるアルキルベンゾエート化合物0.001~10重量部が添加された熱可塑性エラストマー組成物からなる自動車内外装材を提供することにある。

【0010】

【化3】

(I)

(式中、R<sup>1</sup> および R<sup>2</sup> は各々独立に水素原子、炭素原子数1~8のアルキル基またはシクロアルキル基、または炭素原子数6~12のアリール基、アルキルアリール基またはアリールアルキル基を表し、R<sup>3</sup>は炭素原子数1~30のアルキル基を表す。)

【0011】

【発明の実施の形態】以下、上記要旨をもってなる本発明について詳述する。本発明に用いられる熱可塑性エラストマーとしては、ポリプロピレンやポリエチレンなどのポリオレフィン樹脂をハードセグメントとして、EPDMなどのゴム成分をソフトセグメントとしたポリオレフィン系熱可塑性エラストマー、ポリスチレンをハードセグメントとして、ポリブタジエンやポリイソプレンなどのポリジエンをソフトセグメントとして用いたブロックコポリマーのポリスチレン系熱可塑性エラストマーなどが挙げられる。また、必要に応じてこれらポリオレフィン系熱可塑性エラストマーにポリスチレン系熱可塑性エラストマーをブレンドしたものでよい。

【0012】熱可塑性エラストマーにおけるソフトセグメントとハードセグメントの形成方法としては、単純ブレンドの他、共重合によるインプラント化および動的架橋に大別できる。また、ポリスチレン系熱可塑性エラストマーの各セグメントの組み合わせとしては、スチレン-ブタジエン-スチレンブロックコポリマー(SBS)、スチレン-イソプレン-スチレンブロックコポリマー(SIS)、スチレン-エチレン-ブチレン-スチレンブロックコポリマー(SEBS)、スチレン-エチレン-プロピレン-スチレンブロックコポリマー(SEPS)およびこれら4種類の水素添加ポリマー、ランダ

ムSBRの水素添加ポリマー(HSBR)およびポリプロピレンとこれらの任意の1以上のポリマーとのブレンド物などが挙げられる。

【0013】一般式(I)におけるR<sup>1</sup>およびR<sup>2</sup>で表されるアルキル基としては、メチル、エチル、プロピル、イソプロピル、ブチル、イソブチル、第二ブチル、第三ブチル、ペンチル、第三ペンチル、ヘキシル、ヘプチル、オクチル、第三オクチルなどが、シクロアルキル基としては、シクロペンチル、シクロヘキシルなどが、アリール基としては、フェニル、ナフチルなどが、アルキルアリールとしては、メチルフェニル、ブチルフェニルなどが、アリールアルキルとしては、フェニルメチル、1-フェニルエチル、クミルなどが挙げられる。

【0014】R<sup>3</sup>で表されるアルキル基としては、メチル、エチル、プロピル、イソプロピル、ブチル、イソブチル、第二ブチル、第三ブチル、ペンチル、第三ペンチル、ヘキシル、ヘプチル、オクチル、第三オクチル、2-エチルヘキシル、ノニル、デシル、ウンデシル、ドデシル、トリデシル、テトラデシル、ペンタデシル、ヘキサデシル、ヘプタデシル、オクタデシル、ペヘニル、トリアコンチルなどが挙げられる。

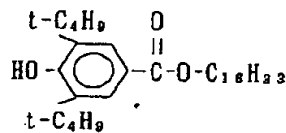
【0015】一般式(I)で表されるアルキルベンゾエート化合物としては、より具体的には、下記の化合物No. 1~No. 5が挙げられる。ただし、本発明は以下

の例示によりなんら制限されるものではない。

【0016】

化合物No. 1

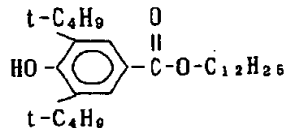
【化4】



【0017】

化合物No. 2

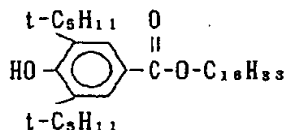
【化5】



【0018】

化合物No. 3

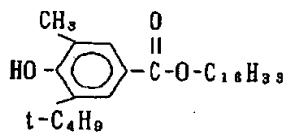
【化6】



【0019】

化合物No. 4

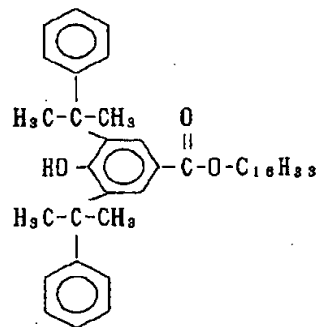
【化7】



【0020】

化合物No. 5

【化8】



【0021】本発明の熱可塑性エラストマー組成物は、前記熱可塑性エラストマー100重量部に前記アルキルベンゾエート化合物0.001~10重量部配合されるものであり、アルキルベンゾエート化合物が0.001重量部未満では自動車内外装材として十分な耐候性が発揮されない。逆に10重量部を超えると昇温時等環境の変化によっては自動車内外装材からのブリードしたりして好ましくない。

【0022】本発明に係る熱可塑性エラストマー組成物は押出成形、射出成形、圧縮成形、積層成形等公知の各手段で適宜自動車内外装材、例えば天井、ドア、座

席、トランク室の内外装材として使用される。

【0023】本発明の熱可塑性エラストマー組成物には、必要に応じてヒンダードアミン系光安定剤を添加することができる。該ヒンダードアミン系光安定剤は耐候性向上の点で、本発明において使用される前記アルキルベンゾエート化合物と相乗効果を奏するので好ましい。また上記組成物には、更に汎用の酸化防止剤等の添加剤を併用することもできる。該酸化防止剤としてはホスファイト系、フェノール系、硫黄系等のものがある。なお、本発明に係る前記以外の紫外線吸収剤も更に併用することができる。

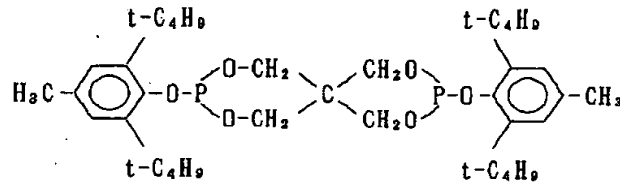
【0024】上記ヒンダードアミン系光安定剤としては、例えば、2, 2, 6, 6-テトラメチル-4-ビペリジルベンゾエート、N-(2, 2, 6, 6-テトラメチル-4-ビペリジル)ドデシルコハク酸イミド、1-[(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオニルオキシエチル]-2, 2, 6, 6-テトラメチル-4-ビペリジル-(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオネート、ビス(2, 2, 6, 6-テトラメチル-4-ビペリジル)セバケート、ビス(1, 2, 2, 6, 6-ペンタメチル-4-ビペリジル)セバケート、ビス(1, 2, 2, 6, 6-ペンタメチル-4-ビペリジル)-2-ブチル-2-(3, 5-ジ第三ブチル-4-ヒドロキシベンジル)マロネート、N, N'-ビス(2, 2, 6, 6-テトラメチル-4-ビペリジル)ヘキサメチレンジアミン、テトラ(2, 2, 6, 6-テトラメチル-4-ビペリジル)ブタンテトラカルボキシレート、ビス(2, 2, 6, 6-テトラメチル-4-ビペリジル)・ジ(トリデシル)ブタンテトラカルボキシレート、ビス(1, 2, 2, 6, 6-ペンタメチル-4-ビペリジル)・ジ(トリデシル)ブタンテトラカルボキシレート、3, 9-ビス〔1, 1-ジメチル-2-(トリス(2, 2, 6, 6-テトラメチル-4-ビペリジールオキシカルボニルオキシ)ブチルカルボニルオキシ)エチル〕-2, 4, 8, 10-テトラオキサスピロ〔5.5〕ウンデカン、3, 9-ビス〔1, 1-ジメチル-2-(トリス(1, 2, 2, 6, 6-ペンタメチル-4-ビペリジールオキシカルボニルオキシ)ブチルカルボニルオキシ)エチル〕-2, 4, 8, 10-テトラオキサスピロ〔5.5〕ウンデカン、1, 5, 8, 12-テトラキス〔4, 6-ビス〔N-(2, 2, 6, 6-テトラメチル-4-ビペリジル)ブチルアミノ〕-1, 3, 5-トリアジン-2-イル〕-1, 5, 8, 12-テトラアザドデカン、1-(2-ヒドロキシエチル)-2, 2, 6, 6-テトラメチル-4-ビペリジノール/コハク酸ジメチル縮合物、2-第三オクチルアミノ-4, 6-ジクロロ-s-トリアジン/N, N'-ビス(2, 2, 6, 6-テトラメチル-4-ビペリジル)ヘキサメチレンジアミン縮合物、N, N'-ビス(2, 2, 6, 6-テトラメチル-4-ビペリジル)ヘキサメチレンジアミン/ジプロモエタン縮合物、2, 2, 6, 6-テトラメチル-4-ヒドロキシビペリジン-N-オキシル、ビス(2, 2, 6, 6-テトラメチル-N-オキシルビペリジン)セバケート、テトラキス(2, 2, 6, 6-テトラメチル-N-オキシルビペリジル)ブタン-1, 2, 3, 4-テトラカルボキシレート、テトラキス(1, 2, 2, 6, 6-ペンタメチル-4-ビペリジル)1, 2, 3, 4-ブタンテ

トラカルボキシレート、3, 9-ビス(1, 1-ジメチル-2-(トリス(2, 2, 6, 6-テトラメチル-N-オキシルビペリジル-4-オキシカルボニル)ブチルカルボニルオキシ)エチル)-2, 4, 8, 10-テトラオキサスピロ〔5.5〕ウンデカン等が挙げられる。

【0025】ホスファイト系酸化防止剤としては、例えばトリスノニルフェニルホスファイト、トリス〔2, 4-ジ第三ブチルフェニル〕ホスファイト、トリス〔2-第三ブチル-4-(3-第三ブチル-4-ヒドロキシ-5-メチルフェニルチオ)-5-メチルフェニル〕ホスファイト、トリデシルホスファイト、オクチルジフェニルホスファイト、ジ(デシル)モノフェニルホスファイト、ジ(トリデシル)ペンタエリスリトールジホスファイト、ジステアリルペンタエリスリトールジホスファイト、ジ(ノニルフェニル)ペンタエリスリトールジホスファイト、ビス(2, 4-ジ第三ブチルフェニル)ペンタエリスリトールジホスファイト、ビス(2, 6-ジ第三ブチル-4-メチルフェニル)ペンタエリスリトールジホスファイト、ビス(2, 4, 6-トリ第三ブチルフェニル)ペンタエリスリトールジホスファイト、ビス(2, 4-ジクミルフェニル)ペンタエリスリトールジホスファイト、テトラ(トリデシル)イソプロピリデンジフェノールジホスファイト、テトラ(トリデシル)-4, 4'-n-ブチリデンビス(2-第三ブチル-5-メチルフェノール)ジホスファイト、ヘキサ(トリデシル)-1, 1, 3-トリス(2-メチル-4-ヒドロキシ-5-第三ブチルフェニル)ブタントリホスファイト、2, 2'-メチレンビス(4, 6-ジ第三ブチルフェニル)-2-エチルヘキシルホスファイト、2, 2'-メチレンビス(4, 6-ジ第三ブチルフェニル)オクタデシルホスファイト、2, 2'-エチリデンビス(4, 6-ジ第三ブチルフェニル)フルオロホスファイト、テトラキス(2, 4-ジ第三ブチルフェニル)ビフェニレンジホスホナイト、9, 10-ジハイドロ-9-オキサ-10-ホスファフェナンスレン-10-オキサイド、トリス〔2-[(2, 4, 8, 10-テトラキス第三ブチルジベンゾ〔d, f〕〔1, 3, 2〕ジオキサホスフェン-6-イル)オキシ]エチル〕アミン、2-エチル-2-ブチルプロピレングリコールと2, 4, 6-トリ第三ブチルフェノールの亜リン酸エステルなどが挙げられる。これらのうち、下記構造式を有するビス(2, 6-ジ第三ブチル-4-メチルフェニル)ペンタエリスリトールジホスファイトが安定化効果に特に優れるので好ましい。

【0026】

【化9】



【0027】また、フェノール系酸化防止剤としては、例えば、2, 6-ジ第三ブチル-p-クレゾール、2, 6-ジフェニル-4-オクタデシロキシフェノール、ステアリル(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)-プロピオネート、ジステアリル(3, 5-ジ第三ブチル-4-ヒドロキシベンジル)ホスホネート、チオジエチレングリコールビス〔(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオネート〕、1, 6-ヘキサメチレンビス〔(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオネート〕、1, 6-ヘキサメチレンビス〔(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオン酸アミド〕、4, 4'-チオビス(6-第三ブチル-m-クレゾール)、2, 2'-メチレンビス(4-メチル-6-第三ブチルフェノール)、2, 2'-メチレンビス(4-エチル-6-第三ブチルフェノール)、ビス〔3, 3'-ビス(4-ヒドロキシ-3-第三ブチルフェニル)ブチリックアシッド〕グリコールエステル、4, 4'-ブチリデンビス(6-第三ブチル-m-クレゾール)、2, 2'-エチリデンビス(4, 6-ジ第三ブチルフェノール)、2, 2'-エチリデンビス(4-第二ブチル-6-第三ブチルフェノール)、1, 1, 3-トリス(2-メチル-4-ヒドロキシ-5-第三ブチルフェニル)ブタン、ビス〔2-第三ブチル-4-メチル-6-(2-ヒドロキシ-3-第三ブチル-5-メチルベンジル)フェニル〕テレフタレート、1, 3, 5-トリス(2, 6-ジメチル-3-ヒドロキシ-4-第三ブチルベンジル)イソシアヌレート、1, 3, 5-トリス(3, 5-ジ第三ブチル-4-ヒドロキシベンジル)イソシアヌレート、1, 3, 5-トリス〔(3, 5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオニルオキシエチル〕イソシアヌレート、テトラキス〔メチレン-3-(3', 5'-ジ第三ブチル-4'-ヒドロキシフェニル)プロピオネート〕メタン、2-第三ブチル-4-メチル-6-(2-アクリロイルオキシ-3-第三ブチル-5-メチルベンジル)フェノール、3, 9-ビス〔1, 1-ジメチル-2-〔(3-第三ブチル-4-ヒドロキシ-5-メチルフェニル)プロピオニルオキシ〕エチル〕-2, 4, 8, 10-テトラオキサスピロ〔5.5〕ウンデカン、トリエチレングリコールビス〔(3-第三ブチル-4-ヒドロ

キシ-5-メチルフェニル)プロピオネート〕等が挙げられる。

【0028】更に、硫黄系酸化防止剤としては、例えばチオジプロピオン酸ジラウリル、ジミリスチル、ジステアリル等のジアルキルチオジプロピオネート類及びペンタエリスリトールテトラ(β-ドデシルメルカプトプロピオネート)等のポリオールのβ-アルキルメルカプトプロピオン酸エステル類が挙げられる。

【0029】また、本発明に係るアルキルベンゾエート系化合物からなる紫外線吸収剤以外の紫外線吸収剤としては、例えば、2, 4-ジヒドロキシベンゾフェノン、2-ヒドロキシ-4-メトキシベンゾフェノン、2-ヒドロキシ-4-オクトキシベンゾフェノン、5, 5'-メチレンビス(2-ヒドロキシ-4-メトキシベンゾフェノン)等の2-ヒドロキシベンゾフェノン類；2-(2'-ヒドロキシ-5'-メチルフェニル)ベンゾトリアゾール、2-(2'-ヒドロキシ-3', 5'-ジ第三ブチルフェニル)ベンゾトリアゾール、2-(2'-ヒドロキシ-3', 5'-ジ第三ブチルフェニル)-5-クロロベンゾトリアゾール、2-(2'-ヒドロキシ-3'-第三ブチル-5'-メチルフェニル)-5-クロロベンゾトリアゾール、2-(2'-ヒドロキシ-5'-第三ブチル-4'-メチルフェニル)ベンゾトリアゾール、2-(2'-ヒドロキシ-3', 5'-ジクミルフェニル)ベンゾトリアゾール、2, 2'-メチレンビス(4-第三ブチル-6-ベンゾトリアゾリル)フェノール等の2-(2'-ヒドロキシフェニル)ベンゾトリアゾール類；フェニルサリシレート、レゾルシノールモノベンゾエート、2, 4-ジ第三ブチルフェニル-3', 5'-ジ第三ブチル-4'-ヒドロキシベンゾエート等のベンゾエート類；2-エチル-2'-エトキシオキサニリド、2-エトキシ-4'-ドデシルオキサニリド等の置換オキサニリド類；エチル-α-シアノ-β, β-ジフェニルアクリレート、メチル-2-シアノ-3-メチル-3-(p-メトキシフェニル)アクリレート等のシアノアクリレート類等が挙げられる。

【0030】本発明の組成物には、更に必要に応じて重金属不活性化剤、造核剤、金属石けん、ハイドロタルサイト類、顔料、有機錫化合物、可塑剤、エポキシ化合物、発泡剤、帯電防止剤、難燃剤、滑剤、加工助剤等をも包含させることができる。

【0031】特に本発明に係る熱可塑性エラストマー組

成物は、顔料を添加してもそのプレートアウトによる加工機への移行がないので、製品の色合わせが容易であり、公知の種々の顔料が使用可能である。本発明の熱可塑性エラストマー組成物に用いられる顔料としては、例えば無機顔料、アゾ系顔料、ニトロ系顔料、アチン系顔料、酸性染料系レーキ顔料、建築染料系顔料、イソインドリノン系顔料、塩基性染料系レーキ顔料、媒染染料系顔料、キナクリドン系顔料、フタロシアニン系顔料、ニトロ系顔料、昼光蛍光顔料、金属粉顔料および重合体結合色素が挙げられる。

【0032】本発明に係る熱可塑性エラストマー組成物の製造の場合、熱可塑性エラストマーへのアルキルベンゾエート化合物、更には他の添加剤の添加方法は特に限定されるものではなく、添加する際の添加剤の形態は粉末でもエマルジョンやサスペンションなどの水分散体でもよく、有機溶剤により溶液としての添加でもよい。また、ブレンダーの種類については特に限定されないが、粉末の場合はリボンミキサー、ヘンシェルミキサー等によるドライブレンド、または単軸もしくは二軸押出機による混練が好都合であり、水分散体とか溶液の場合は通常の縦型ミキサーの使用で充分である。

【0033】さらに、添加される添加剤の形態に応じて添加工程も種々の態様が可能であり、例えば添加剤が粉体の場合は、熱可塑性エラストマー組成物の成形加工時に添加してもよく、また該成形加工後に成形加工品を添加剤の溶液中に浸して吸着または含浸させてもよい。該加工時の添加方法においては、高濃度に配合したマスターバッチを調製したり、顆粒化して粉塵を抑制した後に最終的に目的とする配合において混練されてもよい。

【0034】

【実施例】次に本発明を実施例によって具体的に説明するが、本発明は以下の実施例によりなんら制限されるものではない。

(実施例1) プロピレン樹脂60重量部、EPR20重量部、タルク20重量部、酸化チタン2重量部、ステアリン酸カルシウム0.05重量部、ステアリル(3,5-ジ第三ブチル-4-ヒドロキシフェニル)プロピオネート0.1重量部、トリス(2,4-ジ第三ブチルフェニル)ホスファイト0.05重量部、テトラキス(1,2,2,6,6-ペンタメチル-4-ビペリジル)1,2,3,4-ブタンテトラカルボキシレート0.15重量部およびアルキルベンゾエート化合物0.1重量部をリボンミキサーによりブレンドし、ペレタイザーに供給して250℃で押出加工してペレットを作成した。次いで、250℃で射出成型して、厚さ2mmのシートを作成し、このシートについてサンシャインウエザオメーターを用いて、ブラックパネル温度83℃、120分中18分間の降雨サイクルで、クラックの発生時間及び480時間後の黄色度により耐候性の評価を行った。また、25mm×50mm×2mmの試験片10枚を80℃のオーブン中に置き、1週間後の表面観察によりブルームの有無と程度を観察し、ブルーム無しを○、全面にブルームが認められるものを×として目視で評価した。更に、試験片25gを入れた試験管にカバーガラスで蓋をし、100℃オイルバスで48時間加熱した後、該蓋への付着物の有無と程度(フォギング)をブルームと同様に評価した。これらの結果は表-1に示す。

【0035】(比較例1) 実施例1におけるアルキルベンゾエート化合物を使用しない場合(比較例1-1)、該化合物に代えてアリールベンゾエート化合物の紫外線吸収剤を使用した場合(比較例1-2)及び該化合物に代えてベンゾエート化合物以外の紫外線吸収剤を使用した場合(比較例1-3)について実施例1に準じて評価した。これらの結果は表-1に示す。

【0036】

【表1】

表-1

	紫外線吸収剤	クラック発生時間(hr)	黄色度	ブルーム	フォギング
実施例1	化合物No.1	2900	2.3	○	○
比較例1-1	なし	1000	5.1	○	○
-2	比較化合物1*	2100	12.2	×	×
-3	比較化合物2**	1100	4.5	×	×

\*1: 3,5-ジ第三ブチル-4-ヒドロキシ安息香酸-2,4-ジ第三ブチルフェニルエステル

\*2: 2-(2'-ヒドロキシ-5'-メチルフェニル)-5-クロロベンゾトリアゾール

【0037】(実施例2) ポリプロピレン65重量部、EPDM15重量部、タルク20重量部、酸化チタン2重量部、フタロシアニンブルー1重量部、ハイドロタルサイト0.1重量部、テトラキス[メチレン-3-(3',5'-ジ第三ブチル-4'-ヒドロキシフェニル)プロピオネート]メタン0.1重量部、ビス(2,6-ジ第三ブチル-4-メチルフェニル)ペンタエリスリトールジホスファイト0.1重量部、3,9-ビス[1,1-ジメチル-2-(トリス(1,2,2,6,6-ペンタメチル-4-ビペリジールオキシカルボニルオ

ル)プロピオネート]メタン0.1重量部、ビス(2,6-ジ第三ブチル-4-メチルフェニル)ペンタエリスリトールジホスファイト0.1重量部、3,9-ビス[1,1-ジメチル-2-(トリス(1,2,2,6,6-ペンタメチル-4-ビペリジールオキシカルボニルオ

キシ)ブチルカルボニルオキシ)エチル]-2,4,8,10-テトラオキサスピロ〔5,5〕ウンデカン0.2重量部及びアルキルベンゾエート0.3重量部を実施例1と同様の方法でペレットにし、該ペレットを用いて240℃の混練ロールで厚さ0.1mmのシートに成形した。得られたシートから25mm×50mm×0.1mmの試験片を作成し、このシートについてサンシャインウェザオメーターを用いて実施例1と同様の条件下に、シート表面にクラックの発生するまでの時間を測定して耐候性試験の評価を行った。また、実施例1と同様にしてブルームおよびフォギングを評価した。その

結果を表-2に示す。

【0038】(比較例2)実施例2におけるアルキルベンゾエート化合物を使用しない場合(比較例2-1)、該化合物に代えてアリールベンゾエート化合物の紫外線吸収剤を使用した場合(比較例2-2)及び該化合物に代えてベンゾエート化合物以外の紫外線吸収剤を使用した場合(比較例2-3)について実施例2に準じて評価した。これらの結果は表-2に示す。

【0039】

【表2】

表-2

	紫外線吸収剤	クラック発生時間(hr)	ブルーム	フォギング
実施例2	化合物No.1	2700	○	○
比較例2-1	なし	800	○	○
-2	比較化合物1 <sup>*1</sup>	1000	×	×
-3	比較化合物2 <sup>*2</sup>	1800	×	×

\*1, \*2: 表-1に示した化合物と同じ。

【0040】(実施例3)プロピレン-エチレン共重合体70重量部、SBS30重量部、フタロシアニンプール1重量部、カルシウムステアレート0.2重量部、テトラキス〔メチレン-3-(3',5'-ジ第三ブチル-4'-ヒドロキシフェニル)プロピオネート〕メタン0.2重量部、ビス(2,6-ジ第三ブチル-4-メチルフェニル)ペンタエリスリトールジホスファイト0.2重量部、ビス(2,2,6,6-テトラメチル-4-ピペリジル)セバケート0.3重量部およびアルキルベンゾエート化合物0.3重量部を実施例2と同様に配合してペレット化し、これを用いて240℃混練ロールにより混練し厚さ0.1mmのフィルムを作成した。このフィルムから実施例2と同様にして試験片を切り取り、降雨サイクルを行わない以外は実施例1と同様にしてサンシャインウェザオメーターによる処理をして、脆化時間を耐候性として評価し、2000時間後の黄色度を耐

候着色性として評価した。また、添加剤による顔料のロールへの移行の程度をプレートアウトとして評価した。具体的には、上記配合物のロール成形後に引き続き、白色顔料配合の塩化ビニル樹脂でシートをロール成形して、塩化ビニル樹脂シートが明瞭に青色に着色したものを×、若干着色したものを△、着色が認められなかったものを○として目視評価した。その結果を表-3に示す。

【0041】(比較例3)実施例3におけるアルキルベンゾエート化合物に代えてアリールベンゾエート化合物の紫外線吸収剤を使用した場合(比較例3-1)及び該化合物に代えてベンゾエート化合物以外の紫外線吸収剤を使用した場合(比較例3-2)について評価した。これらの結果は表-3に示す。

【0042】

【表3】

表-3

	試料化合物	脆化時間(hr) (耐候性)	プレートアウト	黄色度 (耐候着色性)
実施例3	化合物No.1	3300	○	4.6
比較例3-1	比較化合物1 <sup>*1</sup>	2300	△	15.6
-2	比較化合物2 <sup>*2</sup>	1500	×	10.8

\*1, \*2: 表-1に示した化合物と同じ。

【0043】(実施例4)プロピレン樹脂60重量部、EPR20重量部、タルク20重量部、酸化チタン2重量部、ステアリン酸カルシウム0.05重量部、テトラキス〔メチレン-3-(3',5'-ジ第三ブチル-

4'-ヒドロキシフェニル)プロピオネート〕メタン0.1重量部、テトラキス(1,2,2,6,6-ペンタメチル-4-ピペリジル)1,2,3,4-ブタンテトラカルボキシレート0.15重量部およびアルキルベ

ンゾエート化合物（化合物No. 1）0.2重量部のほか各種添加剤（表-4）0.1重量部を配合し、250℃で押出加工してペレットを作成した。次いで250℃で射出成形して厚さ2mmのシートを得、黄色度を測定した。このシートについて、降雨サイクルを行わない以外

表-4

	添 加 剤	クラック 発生時間(hr)	黄色度
実施例4-1	な し	1400	10.2
-2	2112**	1400	8.2
-3	HP-10**	1600	5.1
-4	PEP-36**	2200	3.0

\*3：トリス（2，4-ジ第三ブチルフェニル）ホスファイト

\*4：2，2'-メチレンビス（4，6-ジ第三ブチルフェニル）  
-2-エチルヘキシルホスファイト

\*5：ビス（2，6-ジ第三ブチル-4-メチルフェニル）  
ペンタエリスリトールジホスファイト

【0045】

【発明の効果】上記詳細な説明、特に各実施例の結果から明らかなように、熱可塑性エラストマー樹脂の耐候性を改善するに際して、本発明の特定の紫外線吸収剤アルキルベンゾエート化合物を用いた場合には、これに代えて各種酸化防止剤、ヒンダードアミン系光安定剤または

は実施例1と同様にしてサンシャインウェザオメーターによる処理をして、クラックの発生時間を耐候性として評価した。その結果を表-4に示す。

【0044】

【表4】

他の紫外線吸収剤等を用いた場合と比較して耐候性の安定化効果が著しく大きく、しかもブルームやフォギングの問題がないばかりか、加工時のプレートアウトがなく、また顔料を用いた場合にも顔料の移行による加工機汚染がないこと等各種効果が大きいことがわかる。

フロントページの続き

(51)Int. Cl.<sup>6</sup>

C08L 23/16

識別記号

FI

C08L 23/16